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DEVELOPMENT OF A MOLTEN CARBONATE PROCESS FOR REMOVAL OF SULFUR DIOXIDE FROM POWER PLANT STACK GASES

PROGRESS REPORT NUMBER 2
PART IV. CONTRACTOR DEVELOPMENT
April 1, 1968 to October 27, 1968

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SUMMARY

Gas-liquid chemical absorption reactors were studied for application to Atomics International's Molten Carbonate Process. Exploratory experiments showed that absorption of sulfur dioxide in a eutectic mixture of lithium, potassium, and sodium carbonates is very rapid. A continuous wetted-wall absorber demonstrated that the rate of sulfur dioxide absorption in carbonate is limited by mass transfer in the gas phase.

To demonstrate that high efficiency of sulfur dioxide absorption is possible in a continuous reactor, a 2 in. ID column fitted with baffles was assembled. The baffles created the necessary turbulence to allow virtually complete sulfur dioxide removal. The transfer unit height (HTU) of the baffled column was found to be 7 in. compared with approximately 70 in. required for a sulfur dioxide HTU in an unbaffled column. The absorption of nitric oxide (NO) was also tested with the baffle column. Experimental results indicated that nitric oxide reacts slowly in the carbonate melt. The transfer unit height for nitric oxide ranged from 38 to 111 in. Nitric oxide did not significantly alter the absorption of sulfur dioxide.

Sulfur dioxide was absorbed in a spray of molten carbonate under three conditions: low-velocity gas flow (<2 ft/sec), low-velocity gas flow with fly ash added to the sprayed carbonate, and high-velocity gas flow (≈25 ft/sec).

In the low-velocity tests, the percent of sulfur dioxide removed from simulated flue gas varied with the fineness of the spray and with the amount of carbonate in excess of the stoichiometric requirement. Mist-like sprays allowed 99.9+% sulfur dioxide removal.

Fly ash in concentrations up to 0.21 wt % in the carbonate melt had no effect upon the absorption. However, fly ash plugged up all nozzles except for the AI pneumatic nozzle which had large openings, indicating the necessity to filter a recycled carbonate stream.

In the high-velocity absorption tests complete removal of sulfur dioxide was achieved. Although there was only an 18-in. section of pipe above the nozzle and no mist collector, the average loss of carbonate as a mist averaged less than 4 wt % .

Theoretical analysis of liquid diffusion within droplets gave limits for degree of carbonate reaction in various-sized droplets. Analysis of gas-phase diffusion to an aerosol of droplets revealed that sulfur dioxide absorption occurs principally in the vicinity of the spray nozzle where droplet velocity relative to the gas is highest and droplet velocity relative to the absorber walls is decreasing.

The results of experiments and theoretical studies suggest employing a spray reactor for pilot-plant tests. The spray nozzles would be most effective when spraying liquid downward into an upward flowing gas stream. A short honeycomb section of cells would aid in mist collection and contribute a wetted-wall effect to the absorption.

I. INTRODUCTION

Several different gas-liquid reactors are required in the Molten Carbonate Process for removing sulfur dioxide from flue gases. They differ in performance because of differences in the various chemical reactions, the reaction rates being mass-transfer controlled, kinetically controlled, and equilibrium controlled.

The key element of the Molten Carbonate process is the absorber in which SO_2, the flue gas contaminant, is reacted at 450°C with the eutectic mixture of Li_2CO_3, K_2CO_3, and Na_2CO_3, identified as M_2CO_3. The absorption reaction is:

$$M_2CO_3(\ell) + SO_2(g) \xrightarrow{450°C} M_2SO_3(\ell) + CO_2(g) \qquad (1)$$

The metal sulfite rapidly disproportionates,

$$4M_2SO_3(\ell) \longrightarrow 3M_2SO_4(\ell) + M_2S(\ell) \qquad (2)$$

with a half life of several minutes as the absorber effluent is heated above 600°C for reduction.

After suspended solids (fly ash) are removed from the melt, the carbonate-sulfate-sulfide mixture enters the reduction reactor where the sulfate is reduced with a suitable reducing agent, for example hydrogen by the reaction

$$M_2SO_4(\ell) + 4H_2(g) \xrightarrow[\text{Metal Catalyst}]{600°C,\ 10\ atm} M_2S(\ell) + 4H_2O \qquad (3a)$$

or with carbon by

$$M_2SO_4(\ell) + 2C(s) \xrightarrow{800°C,\ 1\ atm} M_2S(\ell) + 2CO_2(g) \qquad (3b)$$

Finally, the metal sulfide is reacted with water and carbon dioxide to allow regeneration of the metal carbonate and provide hydrogen sulfide for production of sulfur, a salable product.

The regeneration reaction is

$$M_2S(\ell) + CO_2(g) + H_2O(g) \xrightarrow{425°C,\ 1\ atm} M_2CO_3(\ell) + H_2S(g) \quad (4)$$

This Progress Report is concerned with only the absorption process (Reaction 1). The purpose of the study was to define the fundamental properties of sulfur dioxide absorption in carbonates, evaluate various gas-liquid reactors and components, and interpret the test results for design of a pilot plant reactor. Preliminary studies were carried out in small absorption columns of the wetted-wall and baffle types. Absorption under conditions similar to those in a power plant was studied in a spray absorber with various nozzles at low gas velocities with pure and fly-ash-containing carbonate melt. Later absorption was investigated in gas flow at 25 ft/sec for study of mist entrainment. Theoretical analyses of diffusion within spray droplets and sulfur dioxide mass transfer from a gas to a gas-aerosol interface were applied to a molten carbonate spray absorber.

II. THE SULFUR DIOXIDE ABSORPTION REACTOR

A. PRELIMINARY STUDIES

Preliminary studies showed the feasibility of using molten alkali-metal carbonates to absorb SO_2 from flue gases. (Grantham and Heredy, 1966).[1] A mixture of 1 to 20% sulfur dioxide in carbon dioxide was bubbled through 1 in. of M_2CO_3 melt at 500°C. This simple contacting system allowed only a fraction of a second contacting time, yet caused 99.9+% absorption of the sulfur dioxide, with sulfite product concentration as high as 30 mole %. These results indicated that the absorption of sulfur dioxide in molten carbonates is indeed very rapid under a wide range of operating conditions.

The first test of absorption in a modeled commercial reactor was carried out in a wetted-wall column with continuous gas and molten salt flows. The wetted-wall contactor was made of a 24-in. long section of 1-in. diameter type 347 stainless steel tube, provided with a melt distribution weir at the top and a gas inlet and melt outlet at the bottom. Tests were performed with this contactor primarily to determine if the mass transfer in scrubber contactors was gas-film limited or not, and also to measure the mass transfer coefficient. A total of 32 tests were carried out, over the range of operating conditions listed below:

Gas Flow Rate	25-250 scfh
Gas Velocity	2.75-30.8 ft/sec
Reynolds Number	580-5360
Gas Temperature	660-850°F
Salt Flow Rate	1-6 lb/hr
Salt Temperature	840-900°F
Inlet SO_2 Concentration	0.1-0.61 vol %

During each test, the sulfur dioxide concentration in the inlet and exit gas streams was measured with a gas chromatograph, and the results used to calculate the mass transfer coefficient for the column.

The test results were quite scattered; however, the Gilliland-Sherwood Equation[2] for wetted-wall columns fell within the scatter, indicating that the absorption rate of sulfur dioxide in molten carbonate is gas-film limited. These tests, however, served only to show the mass transfer mechanism of absorption: a wetted-wall column large enough for a commercial power plant would not be practical. The test data and a more detailed discussion of the results are given in the first summary report, reference 3.

With the demonstration of very rapid chemical reaction and verification that the absorption process is gas-phase-resistance controlled, a baffle column and spray absorber were assembled and studied to aid in devising a pilot plant absorber.

B. THE BAFFLE COLUMN ABSORPTION REACTOR

1. Purpose of the Experiments

Because of size limitations, the wetted-wall column was at best able to remove only 70% of the sulfur dioxide from the gas inlet stream. In order to demonstrate 90% or better sulfur dioxide removal in a continuous flow reactor, a baffle column absorber was designed and tested. The baffle column was also utilized to test nitric oxide removal by carbonates and the influence of nitric oxide upon sulfur dioxide absorption.

2. Apparatus and Procedure

The column is shown in Figure 1. The heart of the baffle column absorber was a 2-in. I.D. stainless steel tube fitted with 9 baffles, segment shaped, spaced 3-in. apart, and inclined at 15° from horizontal. Each baffle extended across three-quarters of the column's diameter and blocked 80.5% of the column cross-section area.

Above the column was a heated melt tank containing molten carbonate. Molten carbonate flowed from the tank, through a valve, into the top plate of the column. The melt trickled through the column and left through a tube at the bottom. All parts of the column, tank, and pipes were heated externally with electrical heaters.

In several tests nitrogen oxide, NO, was added to the inlet gas stream to determine its effect upon sulfur dioxide removal as well as the ability of carbonate to scrub nitric oxide from flue gas.

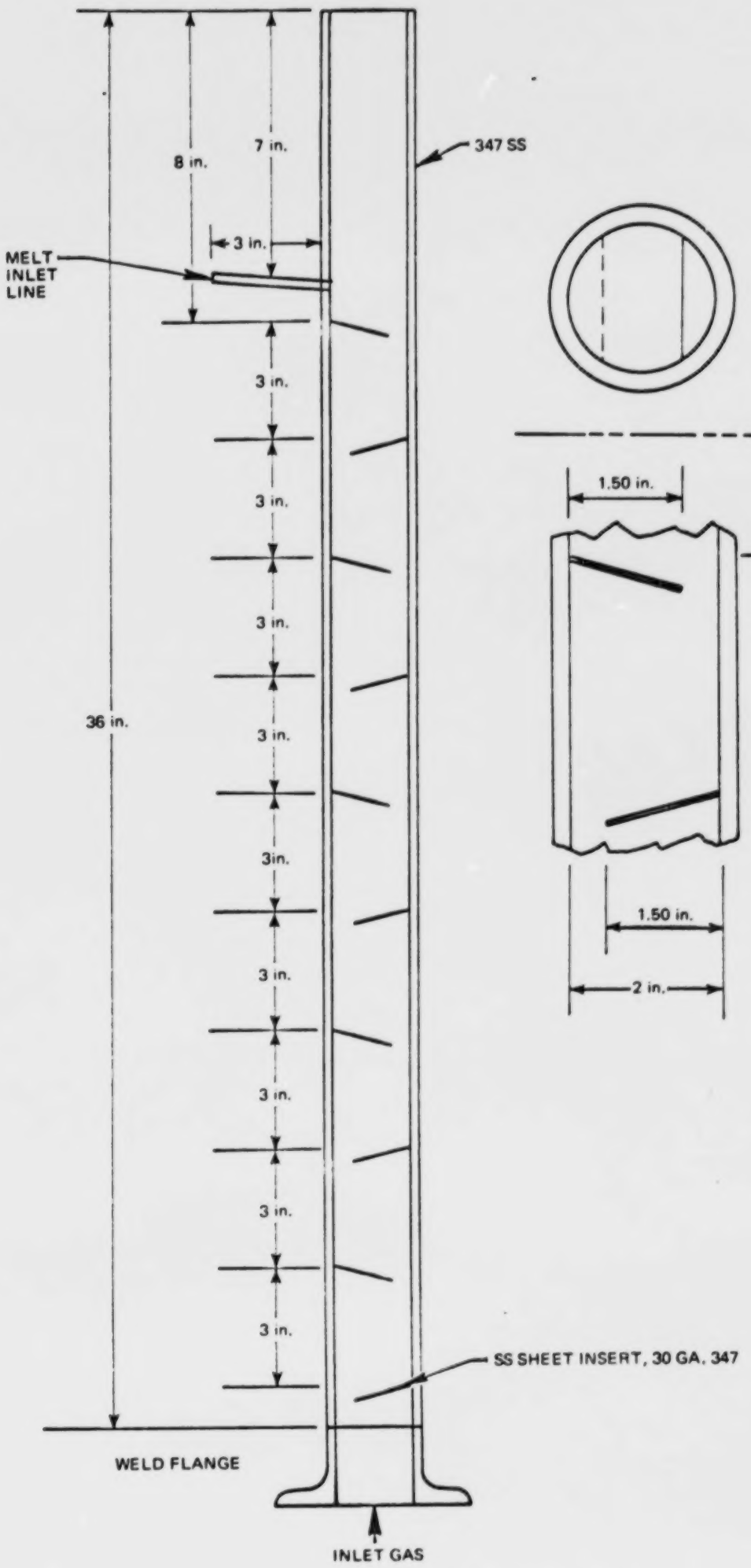

Figure 1. Construction of Baffle Column Absorber

Sulfur dioxide, nitric oxide, and carbon dioxide from tanks were metered through flowmeters, mixed, heated in a tube furnace, and the mixture was introduced at the bottom of the column. Gases exiting at the top of the column were vented to a vacuum hood. Sulfur dioxide and nitric oxide concentration in inlet and outlet gas streams were measured with gas chromatographs.

3. Results

The experimental results and some reduction of the data are listed in Table 1.

4. Discussion

A wetted-wall column operating in laminar or transition flow will absorb gases less effectively than predicted by the Gilliland-Sherwood equation

$$\frac{kD}{c\mathscr{D}} = 0.023 \left(\frac{Dv}{\nu}\right)^{0.83} \left(\frac{\nu}{\mathscr{D}}\right)^{0.44} \qquad (5)$$

or

$$N_{SH} = 0.023\, N_{Re}^{0.83}\, N_{Sc}^{0.44} \qquad (6)$$

which applies only to fully turbulent flow. In the above equations, the symbols are defined as

k = mass transfer coefficient (moles/cm^2 sec)

D = column diameter (cm)

c = molar density of gas (moles/cm^3)

$\mathscr{D}$ = diffusivity (cm^2/sec)

ν = kinematic viscosity (cm^2/sec)

v = gas velocity (cm/sec)

$N_{sh} = \frac{kD}{c\mathscr{D}}$, Sherwood number (dimensionless)

$N_{Re} = \frac{Dv}{\nu}$, Reynolds number (dimensionless)

Table 1. Test Results with the Baffle Column for SO_2 Absorption

Gas Flow Rate			Temp	ΔP	Gas Concentrations, ppm						% Removal		Reynolds No.	HTU's (Inches)		
					Calculated		Chromatograph									
					Inlet		Inlet		Outlet							
CO_2	SO_2	NO	(°F)	in H_2O	SO	NO	SO_2	NO	SO_2	NO	SO_2	NO	**	SO_2	NO	Wetted-Wall
30	0.51	0	805		17000	0	15400	0	1200	0	92		325	10.5		59
30	0.51	0	805	0.14	17000	0	15400	0	800	0	95		325	9.1		59
30	0.254	0	815	0.14	8500	0	9200	0	350	0	96		325	8.3		59
50	0.495	0	900	not meas	9900	0	10000	0	50*	0	99+		540	5.1		64.5
55	0.159	0	940	not meas	2900	0	3600	0	50*	0	99		580	6.3		65
55	0.301	0	900	not meas	5400	0	6200	0	50*	0	99		595	5.6		65
61	0.50	0	900	0.325	8200	0	8200	0	650	0	92		664	10.5		66.5
60	0.254	0	815	0.325	4200	0	4400	0	200	0	95		688	8.7		67
60	0.254	0	815	0.325	4200	0	4400	0	200	0	95		688	8.7		67
120	0.51	0	950	1.7	4200	0	3800	0	50*	0	99+		1270	6.3		74
120	0.51	0	770	1.7	4200	0	3800	0	50*	0	99+		1400	6.3		75.5
120	0.251	0	975	1.7	2100	0	2100	0	50*	0	98+		1280	7.2		74
160	0.51	0	790	4.0	3200	0	2700	0	50*	0	98+		1830	6.8		78
50	0.157	0.024	850	4.0	3140	448	2980	450	50*	220	99.5	51	570	5.4	38	65
50	0.148	0.010	870	4.0	2960	192	2900	220	50*	150	99.3	32	555	5.4	70	65
75	0.197	0.038	885	4.0	2610	510	2400	530	50*	385	99.1	29	830	5.6	84	68
60	0.210	0.069	910	4.0	3500	1150	3540	1340	50*	1050	99.5	22	652	5.2	111	65
60	-	0.039	870	4.0	-	654	-	590	50*	400	-	32	675	-	70	65.5

*SO_2 not detected in exit gas
**Based on unbaffled column. HTU-N_{Re} relation derived in Appendix A

$N_{Sc} = \frac{\nu}{\mathcal{D}}$, Schmidt number (dimensionless)

Accordingly, the HTU* values given in Table 1 for an unbaffled column, where the Reynolds number is less than 2000 are lower limits. Comparison of measured HTU's calculated by the equation

$$HTU = L/\ell n\ (C_{in}/C_{out}),$$

with HTU's predicted for an unbaffled (or wetted-wall) column† indicates the strong effect of the baffles in absorption of SO_2. For example, in the first run (Table 1) the 59 in. required for 63.2% SO_2 reduction in the unbaffled column was reduced to 10.5 in. by adding the baffles. This striking improvement is attributable to the turbulence caused by the baffles. The SO_2-bearing gas is agitated vigorously and brought into intimate contace with the liquid film flowing down the column, thus allowing a greater absorption rate.

However, the improvement in absorption effectiveness comes at a cost of higher pressure drop across the column. While the ΔP in the unbaffled column is negligible, addition of baffles causes a ΔP of up to several inches of water at the higher gas flow rates.

Results of one run showed the column's ability to remove SO_2 to be independent of liquid flow rate. During the run, there was no carbonate flow and all sulfur dioxide removal was from residual, stagnant carbonate in the column. Still, after several minutes operation, no sulfur dioxide could be detected in the exit gas. Other runs had been made with salt flow rates of between 2.3 and 12 lb salt/hr.

---

*The HTU or "height of a transfer unit" is defined as that height of adsorption column in which the concentration of transferred species (SO_2 in the gas) is reduced by $1 - 1/e = 63.2\%$. The HTU is related to the "half-length, Z" or column height in which concentration is reduced by 50% by $HTU = 1.44\ Z_{\frac{1}{2}}$.
†The relation between HTU and Reynolds number is derived in Appendix A

The HTU's measured in the last five runs (Table 1) suggest that nitric oxide concentrations up to 0.1% have no measurable detrimental effect upon sulfur dioxide absorption.

The absorption performance of sulfur dioxide indicates a gas phase HTU of the order of eight inches which would apply to nitric oxide also if it were absorbed as rapidly as sulfur dioxide. The measured HTU's for nitric oxide absorption of 38 to 111 in. (Table 1) indicate that nitric oxide is absorbed sparingly or slowly in the carbonate, significantly less than sulfur dioxide. It follows then that resistance to nitric oxide absorption occurs in the liquid phase because of a slow rate of nitric oxide reaction with the melt. Gas residence time in the baffle column was about three seconds with a velocity averaging 1 ft/sec. The purpose of tests with nitric oxide however, was not to measure its absorption rate, but to determine its effect upon sulfur dioxide absorption. The rate of nitric oxide absorption in actual practice may be improved by the presence of oxygen and sulfides.

5. Conclusions

a. The presence of baffles in a wetted-wall column significantly increased its effectiveness in absorbing gases.

b. The improved gas-absorbing performance is attributable to increased turbulence and, in turn, improved gas-phase mass transfer.

c. Nitric oxide, NO, at concentrations below 0.1% had no measurable effect upon the absorption of sulfur dioxide by molten metal carbonates.

d. Nitric oxide is absorbed slowly by pure carbonate melt.

e. A baffle column would be inadequate for sulfur dioxide removal in a full-scale power plant because of pressure drop limitations.

C. SULFUR DIOXIDE ABSORPTION IN THE SPRAY REACTOR

1. Absorption at Low Velocities

a. Purpose of the Experiments

Several researchers have evaluated and suggested the use of spray contractors in gas-liquid mass transfer applications (Kleinschmidt and Anthony [4], Johnstone and Kleinschmidt [5], Schutte and Koerting Company [6]).

Because of the potentially high specific surface available with a finely divided spray, effective absorption of sulfur dioxide with atomized carbonate melt is possible. Furthermore, spray contacting offers virtually no resistance to the gas flow and the pressure drop in a spray reactor is negligible.

The purpose of these experiments was twofold. The first objective was to evaluate performance of spray nozzles which would atomize molten carbonates under the unusual conditions of high temperature, high liquid surface tension, and severe corrosiveness. The second objective was to test the effectiveness of a spray reactor in removing sulfur dioxide from flue gases.

b. Apparatus and Procedure

(1) The Contactor

The design of the gas liquid contactor (Figure 2) allowed versatility in operation. Liquid supplied at the top could be delivered to a nozzle positioned anywhere in the column. Sulfur-dioxide-bearing gas entered at the bottom. An inlet at the top of the column carried atomizing gas for pneumatic nozzles. The window at the middle of the column gave a view of the nozzle in operation, a feature which proved useful. The column, pipes, and melt tank fabricated from 347 stainless steel pipe were heated with strip heaters welded to their outsides. The entire system was insulated with foil-backed alumina wool.

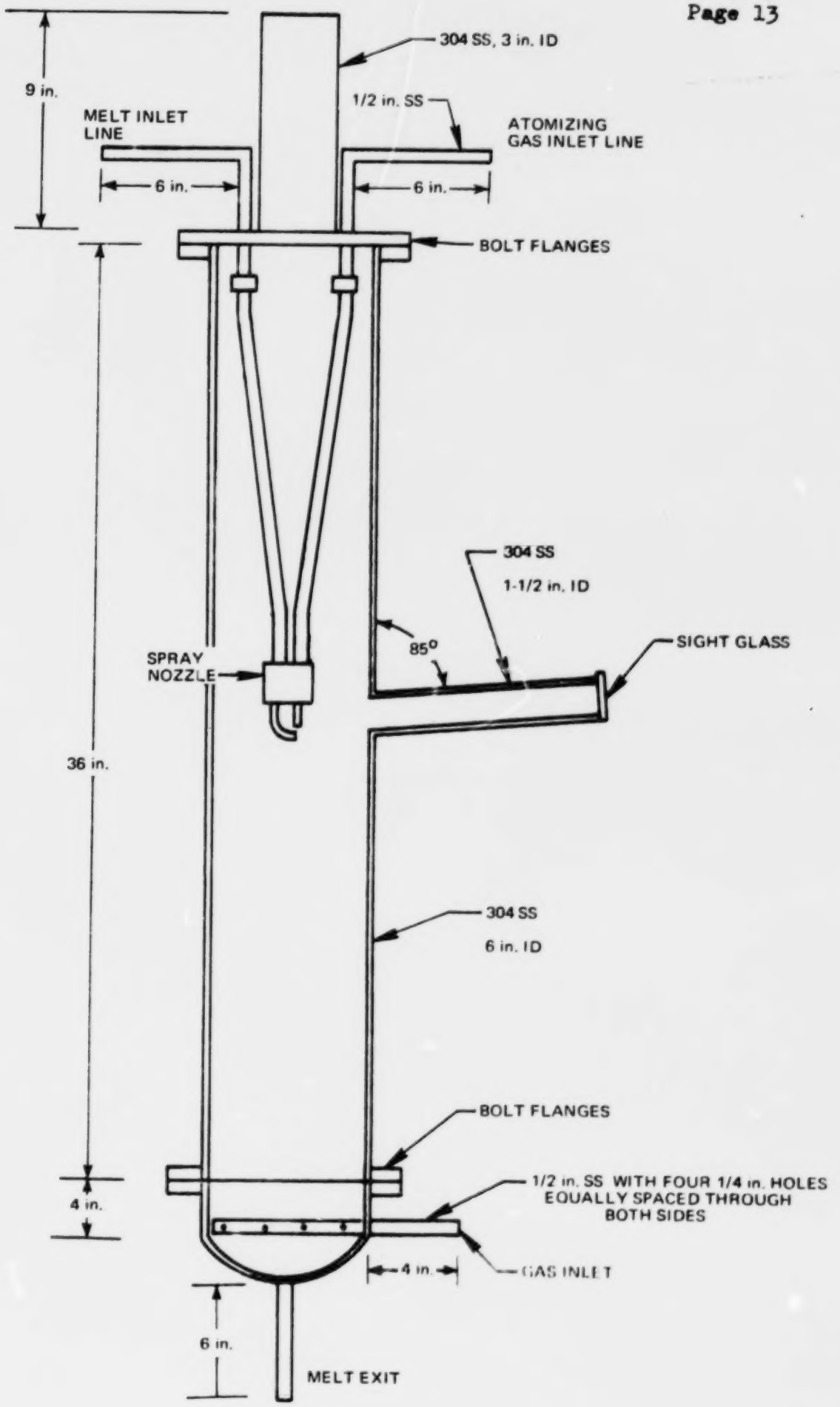

Figure 2. Construction of the Absorption Column

(2) The Nozzles

The nozzles tested were of several types; pneumatic nozzles, whirljet nozzles, an impaction nozzle and an insert-type atomizing nozzle.

The Atomics International pneumatic nozzle was patterned after a bug-spraying nozzle. The nozzle, pictured in Figure 3a, was constructed of stainless steel tubing fitted to a solid stainless steel cylinder. The tips were adjusted using air and water. The formation of droplets relied upon energy of high-speed gas flowing past the liquid tip. Increasing the gas-to-liquid ratio, caused formation of smaller droplets. The feature distinguishing this nozzle from the others was the relatively large diameter of the liquid tubes, so designed to avoid fouling by any solids suspended in the liquid carbonate.

Spraying Systems Company of Bellwood, Illinois, supplied the commercial pneumatic nozzles. The internal mix nozzle (Figure 3b) formed droplets within the nozzle while the external mix nozzle (Figure 3c) utilized two high-speed gas jets to atomize the liquid stream outside of the nozzle.

Whirljet nozzles, supplied by Spraying Systems Co. (Figure 4) rely upon centrifugal force of the fluid to throw it into a hollow cone which breaks into droplets outside the nozzle. The liquid attains a whirling motion inside the nozzle.

An "atomizing" nozzle, supplied by Spraying Systems Co. (Figure 4b) is similar to a whirljet nozzle, however, the spinning motion of the fluid occurs as the fluid passes through helical slots in an insert.

The impact nozzle, fabricated at Atomics International (Figure 4c) operated by impinging a stream of molten salt upon the edge of a target plate.

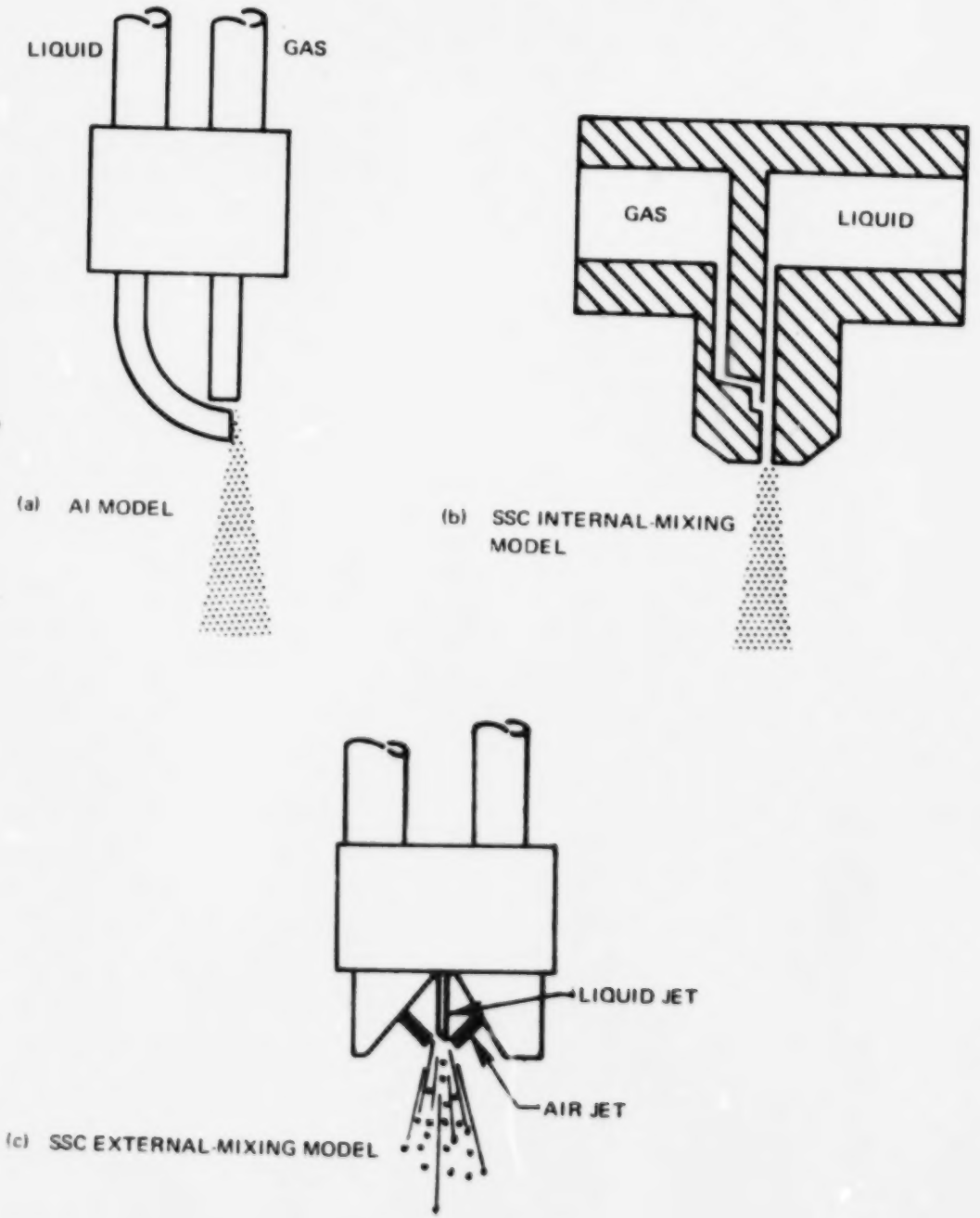

Figure 3. Pneumatic Nozzles

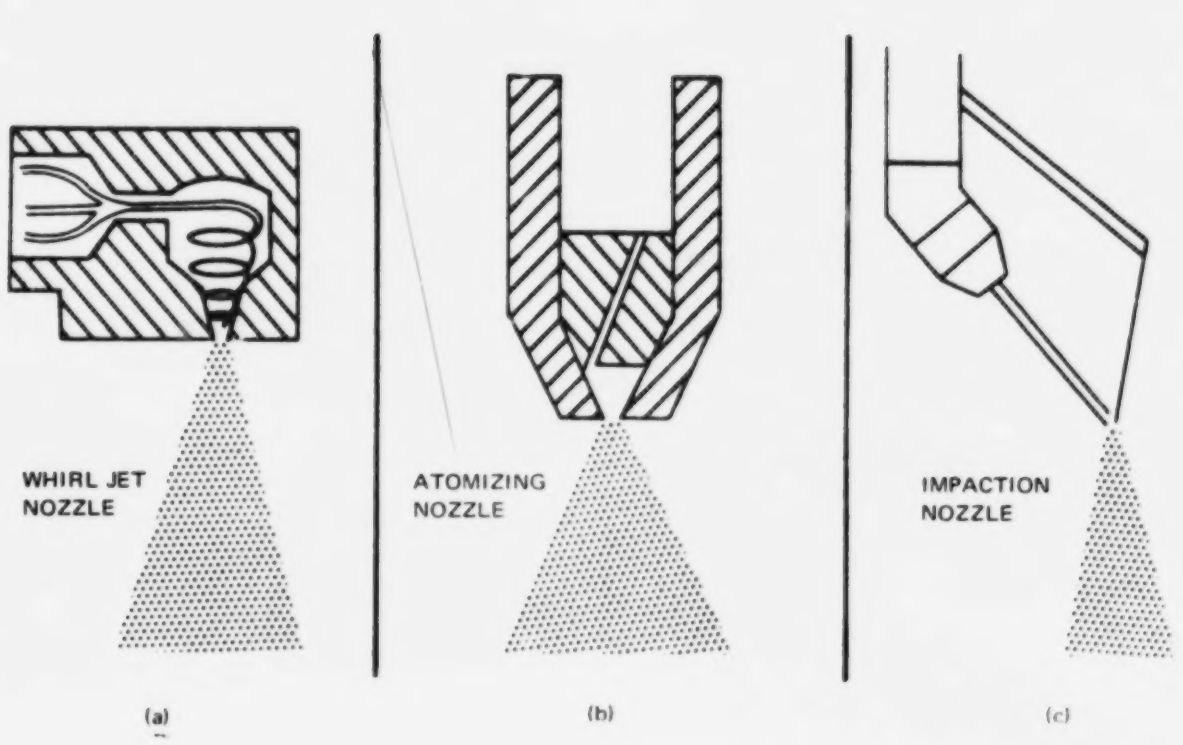

Figure 4. Non Pneumatic Nozzles

(3) Procedure

After the CO_2 and SO_2 flow rates were set, temperatures of the absorber wall and inlet melt were recorded. By watching the spray through the window, the operator set the liquid flow rate to give a "good" spray. Liquid exiting from the bottom of the column was collected for timed intervals and weighed to determine the liquid flow rate through the column.

c. Results

Results of spray absorption tests with various nozzles in the 15 cm I.D. column are listed in Tables 2 through 7 in chronological order. Gas temperatures entered in the table are the averages of inlet and outlet gas stream temperatures. Gas flow rates were read from flow meters and were corrected to the column temperature. Melt flow rates were calculated from the weight of melt leaving the column in a measured time interval. Sulfur dioxide concentration of inlet gas was calculated from inlet gas flow rated or measured with the gas chromatograph. Sulfur dioxide concentration of exiting gas was measured with the chromatograph. Values in the "percent removal" column were calculated from measured values of inlet and outlet concentrations of sulfur dioxide from the formula, percent removal = 100(1 - conc. at outlet/conc. at inlet). The percent of metal carbonate (M_2CO_3)reacted was determined by chemical assay. The remarks column lists observations made during the tests.

d. Discussion

(1) General

In almost all of the tests, the cone of liquid spray from the nozzle intercepted the absorber column walls. Accordingly full use of the liquid spray could not be realized.

Table 2. SO_2 Absorption with the Atomics International Pneumatic Nozzle

23 May	Gas Temp. °F	CO_2 Flow Rate scfh	CO_2 Flow Rate t/min	Mole SO_2 min (G)	Mole M_2CO_3 min (L)	G/L	SO_2 Conc. vol % Inlet	SO_2 Conc. vol % Outlet	%* Removal	$\%M_2CO_3$ Reached
1	1100	453	131	0.025	1.34	0.019	0.60	0.07	98.4+	1.3
2	1100	453	131	0.025	1.34	0.019	0.68	ND	99.24	
3	1000	457	132	0.021	1.34	0.019	0.48	ND	99.8+	
4	1000	386	104	0.046	0.88	0.052	2.83	ND	99.9+	16.3
5	1000	386	104	0.046	0.88	0.052	2.80	ND	99.9+	
6	1000	386	104	0.046	0.88	0.052	2.80	ND	99.9+	
7	1000	386	104	0.046	0.88	0.052	2.80	ND	99.9+	
8	1000	386	104	0.046	0.88	0.052	2.25	ND	99.9+	
9	1000	386	104	0.046	0.88	0.052	2.20	ND	99.9+	
10	1000	386	104	0.046	0.88	0.052	1.65	ND	99.9+	
11	1000	386	104	0.046	0.54	0.085	1.65	ND	99.9+	58.3
12	1000	290	78	0.178	0.54	0.33	3.90	2.10	46.1	
13	1000	290	78	0.178	0.54	0.33	8.2	ND	99.89+	
14	1000	290	78	0.178	0.54	0.33	8.2	0.42	94.9	
15	1000	290	78	0.178	0.54	0.33	8.2	0.61	92.5	
16	1000	290	78	0.168	0.54	0.32	8.15	0.75	90.8	

*% removal where outlet SO_2 was not detected (ND) was calculated on an outlet SO_2 concentration of 20 ppm, the chromatograph's sensitivity.

Table 2. (cont.) SO_2 Absorption with the Atomics International Pneumatic Nozzle

27 May	Gas Temp. (°F)	CO_2 Flow Rate scfh	CO_2 Flow Rate ft/min	Mole SO_2 / min (G)	Mole M_2CO_3 / min (L)	G/L	SO_2 Conc. vol % Inlet	SO_2 Conc. vol % Outlet	% Removal	Remarks
1	830	250	60	0.019	0.29	0.066	0.85	ND	99.9+	Fine Mist
2	860	250	60	0.025	0.29	0.086	2.51	ND	99.9+	Fine Mist
3	860	250	60	0.025	0.29	0.086	2.47	ND	99.9+	Fine Mist
4	860	250	60	0.025	0.17	0.147	3.30	0.30	90.9	Visible
5	860	250	60	0.025	0.17	0.147	2.80	0.21	92.5	raidropos
6	860	250	60	0.025	0.17	0.147	2.80	0.25	91.1	"
7	860	308	74	0.06	0.15	0.40	3.46	0.34	90.2	"
8	860	308	74	0.06	0.15	0.40	3.40	0.35	89.7	"
9	860	308	74	0.06	0.105	0.57	3.40	0.49	85.6	"
10	860	308	74	0.06	0.105	0.57	3.40	0.565	83.4	"
11	860	308	74	0.06	0.255	0.235	3.40	0.35	89.7	"
12	860	308	74	0.06	0.255	0.235	3.40	0.39	88.5	"
13	860	308	74	0.06	0.255	0.235	3.40	0.32	90.5	"
29 May										
14	800	173		0.022	0.20	0.11	0.32	ND	97.4	Frozen salt
15	750	177		0.022	0.11	0.2	0.18	ND	99.0	on nozzle
16	735	209		0.022	0.05	0.44	0.40	0.02	95	Raindrops
17	770	368		0.046	liquid outlet		0.13	0.13	88.5	Drops + mist
18	755	368		0.084	plugged L not		2.12	1.01	52.3	Drops only
19	735	364		0.084	measured		2.12	0.60	71.7	
20	750	373		0.082	"		2.05	lost	lost	Good mist
21	735	585		0.084	"		2.06	0.06	97.1	Good mist
22	770	707		0.082	"		1.18	0.35	70.3	Coarse spray

Table 3. SO_2 Absorption with the Impaction Nozzle

4 June	Gas Temp. (°F)	CO_2 Flow Rate scfh	CO_2 Flow Rate ft.min	Mole SO_2 min (G)	min (L)	G/L	SO_2 Conc, vol % Inlet	SO_2 Conc, vol % Outlet	% Removal	Remarks
1	890	235	59	0.022	14.7	0.0015	0.77			
2	890	235	59	0.022	14.7	0.0015	0.80	0.24	70	Spray on
3	890	235	59	0.022	14.9	0.0015	0.80	0.05	93.8	Spray on
4	890	235	59	0.022	14.9	0.0015	0.80	0.03	96.3	Spray on
5	890	235	59	0.048	14.9	0.0032	2.90	0.48	83.5	Spray on
6	890	235	59	0.048	14.9	0.0032	2.90	0.25	91.5	Spray off
7	890	235	59	0.048	14.9	0.0032	2.90	0.61	79.1	Spray on
8	890	235	59	0.048	14.9	0.0032	2.90	0.28	90.4	Spray off
9	890	235	59	0.048	14.9	0.0032	2.90	0.6	79.4	Spray off
10	890	235	59	0.048	14.9	0.0032	2.90	0.7	76.0	Spray off
11	890	235	59	0.048	14.9	0.0032	2.90	1.0	65.6	Spray on
12	890	235	59	0.048	14.9	0.0032	2.90	0.48	83.5	1 min Spray on
13	890	235	59	0.048	14.9	0.0032	2.90	0.15	94.9	3 min Spray off
14	890	235	59	0.048	14.9	0.0032	2.90	0.23	92.2	Spray off
15	910	423	110	0.022	14.9	0.0032	0.24	0.18	25.0	Off 30 min
16	910	423	110	0.022	14.9	0.0032	0.24	0.175	27.1	Off 30 min
17	910	423	110	0.022	14.9	0.0032	0.24	0.185	22.9	Spray on
18	910	423	110	0.022	14.9	0.0032	0.24	0.06	75.0	Spray on
19	910	423	110	0.022	14.9	0.0032	0.24	0.06	75.0	Spray off
20	910	423	110	0.022	14.9	0.0032	0.24	0.06	75.0	Spray off
21	910	423	110	0.022	14.9	0.0032	0.24	0.06	75.0	Spray off
22	980	423	110	0.086	14.9	0.0032	0.26	1.45	54.1	Spray on
23	980	423	110	0.086	14.9	0.0032	0.26	0.58	77.7	Spray on
24	980	423	110	0.086	14.9	0.0032	0.26	0.7	73.1	Spray on
25	980	423	110	0.086	14.9	0.0032	0.26	0.62	76.2	Spray off
26	940	612	158	0.048	14.9	0.0032	0.477	0.425	91.1	Spray on

Table 4. SO_2 Absorption with the Spraying Systems Co. Internal Mix Pneumatic Nozzle, Spray Setup #11-1/4JCC

6 June	Gas Temp. (°F)	CO_2 Flow Rate scfh	CO_2 Flow Rate ft/min	Mole SO_2 min (G)	Mole M_2CO_3 min (L)		SO_2 Conc. vol % Inlet	SO_2 Conc. vol % Outlet	% Removal	Remarks
1	760	488	110	0.019	0.50	0.038	0.141	0.063	55.3	Nozzle sputtering
2	800	610	142	0.046	0.50	0.092	0.28	0.105	62.5	Coarse drops
3	860	610	142	0.046	0.42	0.110	0.61	0.135	77.8	No mist
4	850	639	156	0.216	0.42	0.515	1.24	0.185	85.0	No mist
5	850	639	156	0.216	0.42	0.515	1.24	0.220	82.3	No mist
6	850	639	156	0.216	0.42	0.515	1.24	0.250	79.8	No mist
7	850	639	156	0.216	0.42	0.515	1.24	0.295	76.2	No mist
8	850	639	156	0.216	0.42	0.515	1.24	0.265	78.6	No mist
9	850	639	156	0.216	0.42	0.515	1.24	0.315	74.6	No mist
10	800	564	131	0.084	0.60	0.140	0.55	0.120	78.2	No mist
11	800	564	131	0.084	0.60	0.140	0.55	0.150	72.7	No mist
12	800	564	131	0.084	0.60	0.140	0.55	0.155	71.8	No mist
13	800	482	112	0.084	1.23	0.068	0.64	0.145	77.4	Fine rain
14	800	482	112	0.084	1.23	0.068	0.64	0.090	85.9	Fine mist
15	800	482	112	0.084	1.23	0.068	0.64	0.02	96.9	Mist out top
16	800	482	112	0.084	1.23	0.068	0.64	0.01	99.0	Mist out top
17	800	482	112	0.084	1.23	0.068	0.64	0.00	99.6+	Mist out top
18	800	482	112	0.084	1.23	0.068	0.64	0.00	99.6+	Mist out top
19	800	565	132	0.302	1.23	0.246	1.96	0.00	99.6+	Mist out top
20	800	565	132	0.302	1.23	0.246	1.96	0.265	86.4	No mist
21	800	565	132	0.302	1.23	0.246	1.96	0.270	86.2	Out top
22	800	565	132	0.302	1.23	0.246	1.96	0.225	88.5	Out top
23	800	565	132	0.302	1.23	0.246	1.96	0.205	89.5	Out top
24	800	565	132	0.302	1.23	0.246	1.96	0.180	85.1	Out top
25	790	521	121	0.172	1.23	0.140	1.21	0.175	85.5	Out top
26	790	521	121	0.172	1.23	0.140	1.21	0.155	92.9	Out top
27	780	458	105	0.210	1.76	0.119	2.18	0.152	93.0	Mist out
28	780	458	105	0.210	1.76	0.119	2.18	0.152	93.0	Mist out chimney
								Avg. =	83.7	

Table 5. SO_2 Absorption with the 1/8A(SS)3 Whirljet Nozzle

June 10 1968	Gas Temp. (°F)	CO_2 Flow		SO_2 Conc, vol %		% Removal	Spray on or off	Remarks
		scfh	ft/min	Inlet	Outlet			
1	735	228	44	0.575	0.032	94.3	on	Liquid leaves nozzle as a solid conical sheet, no drops or holes visible in sheet. Spray angle about 30°. Nozzle pressure = 20 psig
2	735	228	44	0.575	0.021	96.3	off	
3	735	228	44	0.765	0.035	95.4	off	
4	735	228	44	0.765	0.014	98.2	on	
5	735	228	44	0.765	0.012	98.4	off	
6	930	439	98	1.15	0.46	61.6	on	
7	930	439	98	1.20	0.25	79.2	off	
8	930	439	98	1.20	0.32	73.4	off	
9	930	439	98	1.20	0.34	71.6	on	
10	845	467	99	5.0	1.41	70.6	on	
11	845	467	99	4.6	2.25	53.1	off	
12	845	467	99	5.4	1.22	74.6	on	
13	845	467	99	5.4	0.85	82.3	off	
14	845	467	99	5.4	1.58	67.1	on	
15	845	467	99	5.4	1.50	71.2	off	
16	845	467	99	5.4	1.95	63.9	off	
17	845	467	99	5.4	2.3	57.3	off	
18	845	467	99	5.4	2.5	53.6	off	
19	845	467	99	5.4	2.51	53.5	off	
20	845	467	99	5.4	2.6	51.8	off	
21	845	467	99	5.4	4.6	14.8	off	
22	845	467	99	5.4	4.4	18.5	off	
23	845	467	99	5.4	2.48	54.0	on	
24	845	467	99	5.4	1.00	81.5	off	
25	845	467	99	5.4	1.22	77.4	on	Nozzle pressure = 60 psig
26	845	467	99	5.4	0.45	91.7	on	
27	756	694	135	2.0	1.14	33.0	on	
28	756	694	135	2.05	0.45	78.0	off	
29	756	694	135	2.05	0.73	64.4	off	

Table 6. SO_2 Absorption with the Atomizing Nozzle, 1/4 M(SS)26

	8 June	Gas Temp. °F	CO_2 Flow Rate scfh	CO_2 Flow Rate ft/min	Mole SO_2 min (G)	Mole M_2CO_3 min (L)	$\frac{G}{L}$	SO_2 Conc, vol % Inlet	SO_2 Conc, vol % Outlet	% Removal	Remarks
20 psig	1	880	168	42	0.046	15.4	0.003	0.83	0.13	84.3	Spray on
	2	895	168	42	0.046	15.4	0.003	0.83	0.17	79.5	Spray off
	3	895	160	39	0.046	14.2	0.003	0.83	0.02	98.3	Spray on
	4	870	160	39	0.046	14.2	0.003	1.15	0.21	81.8	Spray off
	5	870	165	41	0.046	14.2	0.003	0.92	0.08	91.4	Spray on
	6	870	165	41	0.046	14.2	0.003	0.92	0.16	82.7	Spray off
	7	870	165	41	0.046	14.2	0.003	0.92	0.05	94.7	Spray on
	8	870	165	41	0.046	14.2	0.003	0.92	0.15	83.7	Spray off
	9	870	165	40	0.046	14.2	0.003	1.0	0.05	95.0	Spray on
	10	865	165	40	0.046	14.2	0.003	1.0	0.165	83.5	Spray off
60 psig	11	880	170	42	0.046	42	0.001	0.92	0.004	99.6	Spray on
	12	880	170	42	0.046	42	0.001	0.92	0.17	81.5	Spray off
	13	880	170	42	0.046	42	0.001	0.92	0.006	99.3	Spray on
	14	880	170	42	0.046	42	0.001	0.92	0.14	84.8	Spray off
	15	915	170	43	0.046	40	0.001	1.05	0.008	99.2	Spray on
	16	915	170	43	0.046	40	0.001	1.05	0.155	85.2	Spray off
	17	915	170	43	0.046	40	0.001	1.05	0.06	94.3	Spray on
	18	915	170	43	0.046	40	0.001	1.05	0.13	87.6	Spray off
	19	930	170	44	0.046	40	0.001	1.05	0.008	99.2	Spray on
150 psig	21	930	170	44	0.046	40	0.001	1.05	0.0	99+	Spray on
	22	930	170	44	0.046	40	0.001	1.05	0.125	88.1	Spray off

Table 7. SO_2 Absorption with the External-Mix Pneumatic Nozzle

lb July	Gas Tem °F	CO_2 Flow scfh	CO_2 Flow ft/min	Mole SO_2 / min (g)	Mole M_2CO_3 / min (L)	G/L	SO_2 Conc. vol % inlet	SO_2 Conc. vol % outlet	% Removal of SO_2	Remarks
1	815	180	36.9	0.254	0.37	0.69	7.24	0.26	96.4	Good mist formed at nozzle
2	815	180	36.9	0.254	0.37	0.69	7.24	0.20	97.2	
3	8.5	180	36.9	0.254	1.04	0.24	7.24	0.11	98.5	

Very little of the liquid sprayed into the column exited with the gas stream as an entrained mist at lower gas flow rates. Only a thin fog could be seen in the exiting gas when any entrained liquid was evident. Attempts to collect this mist indicated that the fraction of liquid entrained in the gas was very small, probably less than 0.1%.

Inspection of the absorber walls during spraying revealed a continuous film of liquid on the walls. Therefore, the absorber must have acted as a wetted-wall column in addition to its performance as a spray reactor. In some runs the column was operated alternately with the spray turned on and turned off to separate the wetted-wall and spray effects. The results are discussed in the following sections.

The following topic numbers parallel Table numbers of the preceding results section.

(2) Performance of the Atomics International Pneumatic Nozzle

Three conclusions are evident from a cursory inspection of the sulfur dioxide removal data for the Atomics Internation pneumatic nozzle:

(a) It allowed the most efficient sulfur dioxide removal.

(b) The extent of sulfur dioxide removal was qualitatively related to the droplet size. Large drops or "rain" were associated with poorer sulfur dioxide removal. Good sulfur dioxide removal was associated with a "mist", that is, droplets too small to be seen individually.

(c) The percent of sulfur dioxide removal was related to the G/L ratio, (moles of SO_2)/(mole of M_2CO_3).

As would be expected from mass transfer fundamentals, the greatly increased gas-liquid interface area of the finely divided mist allowed virtually complete absorption of sulfur dioxide. Collection upon the absorber

walls removed essentially all of the mist before it could escape through the absorber chimney. Wall collection of mist is not assured in a much larger absorber with a higher linear gas velocity and a mist eliminator might be necessary.

Were there no mass-transfer resistance in either the gas or liquid phase, 100% sulfur dioxide removal would be expected for G/L, (moles SO_2/min)/(moles M_2CO_3/min),less than unity. For G/L greater than unity, the percent removal would be given by $(G/L)^{-1}$. The influence of mass transfer resistance in the spray column was to hinder contact of sulfur dioxide and carbonate (which must diffuse to the droplet surface) within liquid droplets, thus reducing sulfur dioxide removal. The reduction of percent SO_2 removal with increasing G/L is shown in Figure 5. The value of 87% SO_2 removal at G/L = 0.57 was close to the value of 83.29% SO_2 removal at G/L = ∞ (See Sections (6), (7)) for which the column's wetted-wall effect predominates. The physics of diffusion within a droplet is discussed in Section 4a.

A 400% excess of melt (excess above theoretical amount in the absence of mass transfer resistance) allowed 95% SO_2 removal. A 900% excess of melt was required for 98% SO_2 removal.

(3) Performance of the Impaction Nozzle

The impaction nozzle operated by impinging a fine stream of liquid upon a metal plate. In water tests, the stream intercepted the plate edge and atomized. In the absorption column the liquid M_2CO_3 stream struck near the plate edge and was deflected to a smooth, continuous sheet of liquid.

The absorption column was run alternately with the liquid "spray" on and off. With the spray off, the absorber acted only as a wetted-wall column. The "spray on" and "spray off" data were paired and tested for a difference in percent SO_2 removal. The difference was not significant.

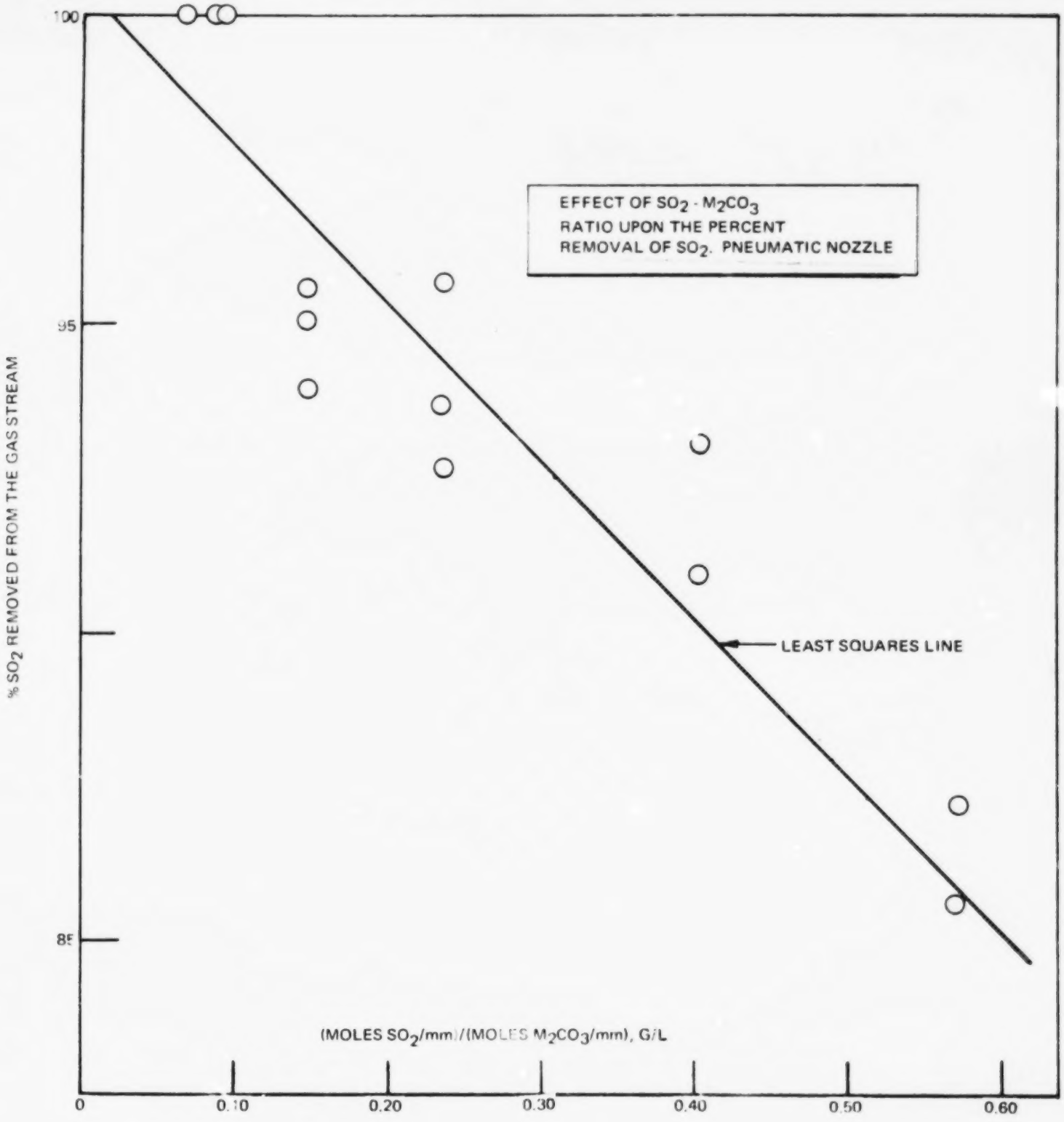
EFFECT OF $SO_2 \cdot M_2CO_3$ RATIO UPON THE PERCENT REMOVAL OF SO_2. PNEUMATIC NOZZLE
100
95
85
% SO_2 REMOVED FROM THE GAS STREAM
LEAST SQUARES LINE
(MOLES SO_2/mm)/(MOLES M_2CO_3/mm), G/L
0
0.10
0.20
0.30
0.40
0.50
0.60

Figure 5.

The mean SO_2 removal for all impaction nozzle runs was 73.7%. Because the liquid flow rate was high (of the order of 1500 gm/min), the nozzle was operated for only five seconds before a gas sample was collected at the column outlet. Later tests showed that a 30-second interval was necessary to allow a representative sample of gas to flow to the chromatograph. Accordingly, the impaction nozzle may have and probably did perform better than indicated in the tests.

(4) Performance of the Spraying Systems Co. Internal-Mix Pneumatic Nozzle

A pneumatic nozzle manufactured by Spraying Systems Company of Bellwood, Illinois was tested in the spray absorber. At times, a smoke-like mist was seen both through the absorber window and emanating from the absorber outlet. Associated with this finely-atomized spray were sulfur dioxide absorption values generally above 90%.

With less atomizing gas supplied to the nozzle, the spray formed into a fine rain or, at times, coarse drops, estimated at 450 microns in diameter. When the mist was absent, sulfur dioxide removal was poorer, being between 55.3-92.9%.

The mean sulfur dioxide removal was found to be 96.8% "with mist" and 79.3% "without mist", a statistically significant difference. This further confirms the effect of droplet size upon rate of sulfur dioxide absorption.

Comparison of the sulfur dioxide-to-melt ratio, G/L, with percent sulfur dioxide removal showed no significant correlation. The effect of drop size probably predominated over the G/L ratio effect.

Results in this test were not influenced by time lags in the chromatograph transmission line (see Section 5) since the pneumatic nozzle was run continuously.

(5) Performance of the Spraying Systems Co. Whirljet Nozzle

The Whirljet nozzle manufactured by Spraying Systems Co. delivered a hollow, conical sheet of liquid with no apparent droplet formation. Higher pressure and flow rate, not attainable with the test system, would have been necessary for droplet formation.

The column was operated alternately with the spray on and spray off for measurement of wetted-wall effect. Comparison of successive runs, in example runs 12, 13, 14, and 15, shows the specious result of more effective absorption with the liquid spray turned off. A search for conditions explaining this anomoly suggested that the interval between the time of turning on the spray and time of outlet gas sampling might influence the observed outlet concentration of sulfur dioxide. It was found that not enough time had been allowed for gas samples collected at the column outlet to travel through several feet of tube to the gas chromatograph. Hence, measured outlet SO_2 concentrations for spray runs were influenced by the previous spray-off run and all spray results with this nozzle are questionable. The mean percent SO_2 removal for "spray-off" or wetted-wall runs was 79.4%

Runs 15 through 22 show the decrease in SO_2 absorption with time when carbonate is not supplied to the column walls. The result of run 23 was evidently influenced by the low concentration of sulfur dioxide still remaining in the chromatograph line from run 22.

(6) Performance of the Spraying Systems Co. Atomizing Nozzle, 1/4 M(SS) 26

This atomizing nozzle (see Figure 4) contains a helically-slotted insert which imparts a spin to liquid leaving the nozzle. The spinning liquid is thrown out in a conical sheet which breaks into a hollow cone of droplets.

A tank pressure of 20 psig was insufficient for satisfactory nozzle performance; the melt left the nozzle in the form of a curved conical sheet. At 60 and 150 psig tank pressure, the nozzle worked satisfactorily. The spray emanated from the nozzle in a cone with droplet formation.

The column was operated alternately with the spray on and off to separate the spray and wetted-wall effects. The results summarized below indicate that the spray contributes significantly above the wetted-wall effect in SO_2 collection.

Average Percent SO_2 Removal

Pressure	Spray on	Spray off	Difference
20 psig	92.74	82.24	10.5 ± 5.8
60 psig	98.32	84.34	14.0 ± 3.4

While the uncertainty is large, the difference in average percent SO_2 removal due to melt tank pressure is statistically significant.

Using the formula

$$\%\ SO_2 \text{ removal by the spray effect} = \frac{\text{total } SO_2 \text{ removal} - WW}{1 - WW}, \qquad (7)$$

where WW is the wetted-wall effect (% SO_2 removal) with the spray off, one obtains values of 59.1 and 89.6 % SO_2 removal by the spray alone for operation at 20 and 60 psig, respectively. The % SO_2 removal attributed to the spray is of the same order of magnitude as that attributed to the wetted-wall.

In most spray tests with the 15 cm ($\frac{1}{2}$ ft) diameter absorber the contribution of wetted-wall-column effect to sulfur dioxide removal was comparable to the spray effect. In a commercial-sized spray reactor, of the order of twenty-five feet in diameter, the HTU would be large, allowing only a fraction of a wetted-wall transfer unit in the absorber length. Wetted wall HTU is related to column diameter by

$$HTU_{ww} \propto D^{1.17} \qquad (8)$$

Hence, increasing the column diameter from $\frac{1}{2}$ foot to 25 feet would increase the wetted-wall HTU over two-hundred fold while the spray HTU would stay constant. Wetted wall effects would therefore be negligible in a large-diameter absorption reactor and spray effects alone would account for removal of sulfur dioxide.

(7) Performance of the Spraying Systems Co. External-Mix Pneumatic Nozzle

The External-Mix, Flat-Spray Pneumatic Nozzle (Figure 3c) was recommended by Spraying Systems Company. The nozzle atomized pure liquid carbonate effectively at a liquid pressure of 20 psig and a gas pressure of 30 to 40 psig. No large droplets were observed. The measured percent removals of sulfur dioxide for three tests were 96.4, 97.2 and 98.5 percent.

Again separating the spray effect from the total observed percent removal of sulfur dioxide (Equation 7), the spray effects for the External Mix pneumatic nozzle were 78.5, 83.2 and 88.0 percent removal of sulfur dioxide.

These values are considerably less than the spray effect of 99+ percent SO_2 removal capable with the AI-developed pneumatic nozzle, but are of the same order of magnitude as the wetted-wall effect.

e. Conclusions

(1) The capability for SO_2 removal improves with decreasing droplet size. The AI pneumatic nozzle which generated a fog-like mist was much more efficient in absorbing SO_2 than other nozzles which produced coarser droplets.

(2) The ratio of gas-to-liquid in the absorption reactor influences sulfur dioxide absorption. Reducing G/L from 0.57 to 0.066 reduced the percent of unreacted sulfur dioxide from 13% to less than 0.1%. It is recommended that the Pilot Plant be designed for a G/L ratio of 0.2 (5 times the stoichiometric carbonate requirement) to allow greater than 95% sulfur dioxide removal.

(3) A commercial pneumatic nozzle of the internal mixing type operated in two modes. When the nozzle generated a mist, SO_2 absorption was high, averaging 96.8%. When the nozzle produced coarser droplets, SO_2 absorption was lower, averaging 79.3%.

(4) The percent SO_2 removal attributable to wetted-wall effects was of the same order of magnitude as the spray effect for medium-performance nozzles (atomizing nozzle and external-mix nozzle). However, in a larger diameter column, wetted-wall effects would be negligible in SO_2 absorption.

(5) Spray droplets are quickly collected upon the walls of a small-diameter (15 cm) column at low gas flow velocities.

2. Sulfur Dioxide Absorption at High Gas Velocities

a. Purpose of the Test

The purpose of this test was to measure the amount of molten carbonate which would be lost as a mist when spraying into a high-velocity stream of flue gas, and to measure the efficiency of sulfur dioxide removal under gas velocity conditions encountered in commercial power plants.

b. Apparatus and Procedure

The absorption column used in low-velocity tests was modified for high velocity flow up to 25 ft/sec. The column diameter was reduced from 6 in. to 4 in. with a pipe insert, flow meters were changed to permit measurement of gas flow up to 3500 scfh, and a new heater was added for heating the gas supplied to the pneumatic nozzle. Nitrogen gas was used for the inert carrier gas rather than carbon dioxide because of its low Joule-Thompson coefficient. Carbon dioxide, used in low-velocity tests, would have cooled on expanding to the point of icing up valves.

In operation, the nitrogen and sulfur dioxide flow rates were set and temperatures were recorded. Molten carbonate was then sprayed into the column. After thirty seconds of operation, a sample of exit gas was collected for chromatographic assay. The gas flow was then shut off to conserve gas.

c. Results

(1) Loss of Carbonate

Two melt tanks full of molten carbonate were sprayed into the column. The weights of carbonate charged to the melt tank and collected at the column's liquid outlet are listed in Table 8.

TABLE 8. Weights of M_2CO_3 Charged to and Recovered from the High-Velocity Absorber

	M_2CO_3 charged to tank	M_2CO_3 recovered at liquid outlet
	kilograms	kilograms
First Tank	18.0	17.0
Second Tank	12.0	12.3

(2) Sulfur Dioxide Removal Efficiency

The results of the sulfur dioxide removal tests are listed in Table 9. The double entries in the column headed "SO_2 (G), mole/min" account for disparity in measuring the sulfur dioxide flow rate. The left-hand entry is the flow rate measured on the rotameter, while right-hand entry is the flow rate obtained from the nitrogen flow rate and the concentration of sulfur dioxide in the nitrogen as measured with the gas chromatograph. These values are carried through the other columns of Table 9. The next to last column gives an estimate of the bounds of a range of removal efficiency values.

d. Discussion

If no melt were lost, the mass of the melt exiting from the absorption reactor would exceed the input mass by an amount ranging between zero and twenty percent, depending on the amount of sulfur dioxide. If all the carbonate (average molecular weight = 100) reacted, only sulfite (of molecular weight = 120) would leave the reactor. How much sulfur dioxide reacted during all the runs is not known. However, the net sulfur dioxide/carbonate ratio is small, and it is estimated that the exiting melt had a molecular weight of about 102. With this assumption, carbonate loss by entrainment in the gas was 7% in the first tankful and -0.5 (gained) in the second tankful.

Table 9. SO_2 Absorption in High Speed Gas Flow with the Atomics International Pneumatic Nozzle

Run No.	Avg. Gas Temp. °F	N_2 Flow Rate scfh	N_2 Flow Rate ft/sec	SO_2 (G) (mole/min)	M_2CO_3 (L) (mole/min)	$\frac{G}{L}$	SO_2 Conc. vol % Inlet	SO_2 Conc. vol % Outlet	% Removal	Remarks
1	735	1270	12.3	0.19 0.85	2.0	0.09 0.42	0.76 3.2	0.32	57.9 90.0	No mist in exit gas
2	735	1270	12.3	0.19 0.85	2.0	0.09 0.42	0.76 3.2	ND	99.7 99.9+	
3	735	1270	12.3	0.19 0.85	2.0	0.09 0.42	0.76 3.2	0.05	93.4 98.4	
4	795	2500	25.3	0.19 0.85	2.0	0.09 0.42	0.39 1.65	0.04	90	
5	790	1340	13.6	0.22 0.98	—	--	0.86 3.62	ND	99.8 99.9+	Mist in exit gas; coarse drops rising near nozzle
6	790	1340	13.6	0.22 0.98	6.6	0.03 0.15	0.88 3.62	ND	99.8 99.9+	
7	790	1340	13.6	0.22 0.98	6.4	0.03 0.15	0.88 3.62	ND	99.8 99.9+	
8	790	1340	13.6	0.22 0.98	5.7	0.04 0.17	0.88 3.62	ND	99.8 99.9+	
9	790	1340	13.6	0.22 0.98	5.7	0.04 0.17	0.88 3.62	0.03	96.5 99.2	
10	790	1340	13.6	0.22 0.98	5.7	0.04 0.17	0.88 3.62	0.03	96.5 99.2	
11	790	1340	13.6	0.22 0.98	5.7	0.04 0.17	0.88 3.62	0.03	96.5 99.2	
12	600	1650	13.9	0.22 0.93	3.0	0.04 0.17	0.88 3.62	0.04	95.4 99.2	
13	600	1650	13.9	0.22 0.93	3.0	0.04 0.17	0.88 3.62	0.04	95.4 99.2	

Table 9 (cont.) SO_2 Absorption in High Speed Gas Flow with the Atomics International Pneumatic Nozzle

Run No.	Avg Gas Temp. °F	N_2 Flow Rate scfh	N_2 Flow Rate ft/sec	SO_2 (G) (mole/min)	M_2CO_3 (L) (mole/min)	$\frac{G}{L}$	SO_2 Conc, vol % Inlet	SO_2 Conc, vol % Outlet	% Removal	Remarks
14	835	2400	25	—	—	—	0.54 —	0.61 —	0	No spray, column acting as a wetted-wall absorber
15	—	—	—	—	—	—	0.54 —	0.15 —	72.2	
16	—	—	—	—	—	—	0.54 —	0.61 —	0	

The loss of carbonate, which is small considering the short absorber length and absence of a demister, is in part explained by the fact that the cone of spray, generated at the nozzle, intercepted the absorber walls. Droplets could be seen impinging upon the walls and upon contact each droplet became part of the wall film; no drops were seen to bounce from the wall. It was evident in watching the spray through the window, that some of the spray was being deflected by the gas and the direction actually reversed in some cases. Figure 6 shows a sketch made during observation through the window. The cone of droplets being blown downward by the atomizing gas probably intercepted the absorber walls. Some of the mist being convected upward escaped through the absorber chimney. The remainder probably contacted the walls and ran down as a film.

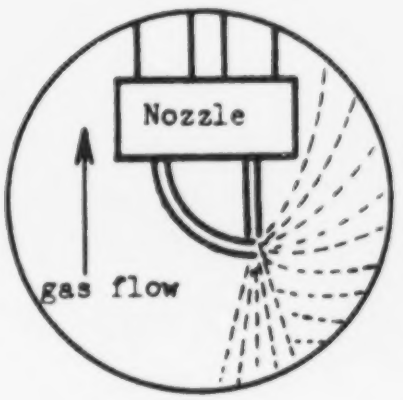

Figure 6. Spray Deflection

The percent of sulfur dioxide absorbed based on gas chromatograph readings exceeded 90% and averaged 98.7%. At the test Reynolds numbers of 6,000 to 13,000, the wetted wall HTU calculated from

$$HTU_{ww} = 11\ D\ N_{Re}^{0.17} \qquad (9)$$

ranged between 190 and 200 in., implying a negligible sulfur dioxide removal by wetted-wall effects. Accordingly the spray accounted for virtually all of the sulfur dioxide removed.

The two main observations in this experiment, (i) that spray droplets are collected upon the walls of a containing tube, and (ii) that a spray is capable of high sulfur dioxide removal efficiency in a high-velocity

gas stream, suggest a design for a pilot plant absorber. The absorption reactor shown in Figure 7, is composed of a sheet metal honeycomb about 10 feet high fitted with downward spraying nozzles near the bottom of each cell.

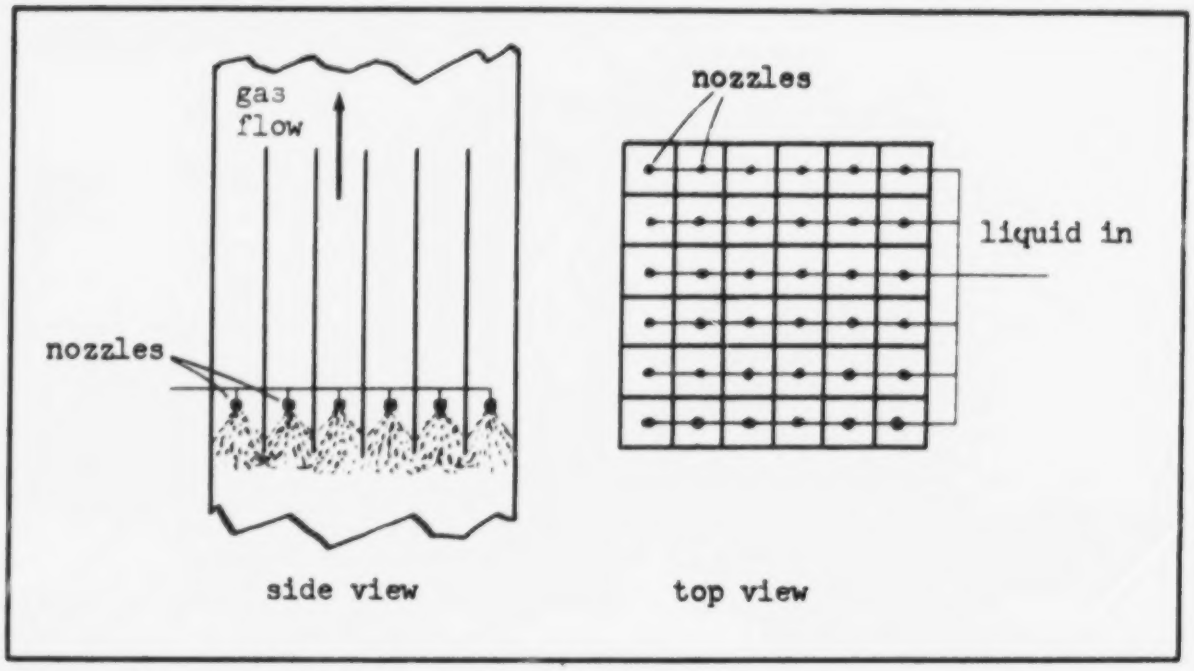

Figure 7. Form of the proposed spray absorber.

Absorption of the sulfur dioxide would occur in the immediate vicinity of the nozzles. The honeycomb section would serve as a spray collector. A mist eliminator would remove the last traces of spray from the gas stream. With honeycomb cells of 1-foot inside diameter, the pressure loss in the honeycomb for a gas flow velocity of 25 ft/sec would be less than 0.1 in. of water.

e. Conclusions

(1) Carbonate loss from atomization within an empty 4 in. I.D. tube, three feet long with a gas velocity of up to 25 ft/sec is less than 7%.

(2) Sulfur dioxide removal efficiency is high; in general, greater than 99% under the conditions listed above for a liquid excess of 6 to 30 times the stoichiometric requirement.

3. Effect of Fly Ash upon Sulfur Dioxide Absorption

a. Introduction

In the Molten Carbonate SO_2 Removal Process, it is estimated that 0.01 to 0.1% weight percent of fly ash will be present in the melt supplied to the absorber. The purpose of the study was to evaluate nozzle performance (SO_2 removal) with fly ash present in the melt.

b. Apparatus and Procedure

The effect of fly ash upon performance of the spray scrubber was studied at low gas velocities (less than 2 ft/sec). Four nozzles were tried; an atomizing (insert type) nozzle, a whirljet nozzle, an external-mix pneumatic nozzle and the pneumatic nozzle developed at AI.

Each nozzle was first tested with pure carbonate melt for ability to spray. Then fly ash was added to the melt tank and spraying was continued. Sulfur dioxide removal performance was measured by comparing the sulfur dioxide concentration at the inlet and outlet of the column.

c. Results and Discussion

The first nozzle evaluated, Spraying Systems Company atomizing nozzle (1/4 in. MSS 26), contained an insert with helical slots. The orifice diameter was 0.086 inches. After an initial spurt of 0.1 wt % ash-in-melt suspension through the nozzle, all flow ceased. Pressure on the melt tank was increased in 20-psig increments to 150 psig to no avail. This nozzle was then removed from the absorption system and cleaned in a weak HCl solution. Detection of fly ash in the cleaning solution was impossible. Many small flakes of rust were shed from the exterior of the nozzle due to the reaction with HCl.

This nozzle was also tested with a suspension of metal carbonate melt containing 0.01 wt % fly ash. At 20 psig pressure, the nozzle sprayed for about one minute and again the flow ceased. The pressure was increased to 60 psig, and the nozzle again sprayed for about one minute before stopping. Upon further increasing the pressure to 120 psig, the nozzle sprayed for about one-half minute, then stopped. Therefore, tests with this nozzle were discontinued.

The next nozzle tested was of the whirljet (no insert) type. The orifice diameter of this nozzle is 0.047 inches. The nozzle could not be made to spray at pressures up to 150 psig with fly ash present in the melt.

At this point, a test was conducted to determine if the plugging was occurring in the nozzles or the associated plumbing. The melt lines were found to be unobstructed.

Two series of runs were made using the pneumatic nozzle developed at Atomics International. The fly ash-melt suspension flowed freely through the pneumatic nozzle. The smallest restriction in the nozzle is about 0.160 inches.

In the first series of tests, a suspension of 0.02 to 0.37 wt % fly ash was sprayed through the AI pneumatic nozzle. The column was operated alternately with the spray turned on and turned off. The sulfur dioxide removal values for 14 observations (7 spray on and 7 spray off) are summarized in Table 10.

TABLE 10 SO_2 Absorption with a Suspension of Fly Ash in Metal Carbonate

Mode of Operation	Spray On	Spray Off	Difference
SO_2 Absorption, %	99.3	91.1	8.2
Associated No. of Transfer Units	5.0	2.4	2.6

The SO_2 absorption attributed to the nozzle alone is that associated with 2.6 transfer units (Table 10) or 92.4% (*). The difference in ability to absorb SO_2 is statistically significant between operation with and without the spray.

The effect of fly ash settling in the melt tank caused initially high concentrations of fly ash to be delivered to the nozzle. The initial concentration was measured at the column melt exit as 0.07 wt %. During the course of the experiment, the fly ash concentration decreased to a final value of 0.025 wt %.

In comparing these results with earlier results from spraying pure carbonate melt, there is no evidence that 0.01 to 0.07 wt % fly ash in the melt affected sulfur dioxide absorption.

The AI pneumatic nozzle was later tested in spraying a more concentrated suspension of fly ash in carbonate melt. The suspension charged to the melt tank was 0.3 wt % in fly ash; however, the cencentration of ash in melt effluent from the column assayed at 0.104 and 0.210 wt % ash in two observations. The mean of three preliminary observations when spraying ash-free melt was 98.4% sulfur dioxide removal. When ash was added to the melt

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* % removal and number of transfer units (N_y) are related by % removal = $100 \left[1 - \exp(-N_y)\right]$

the mean of nine observations was 98.0 sulfur dioxide removal. The difference between 98.0 and 98.4 was not statistically significant at the 95% confidence level.

d. Conclusions

Fly ash suspended in molten M_2CO_3 obstructed flow in nozzles with small openings. The ash-melt suspension flowed freely through a nozzle with openings at least 0.4 cm (0.16 in.) in diameter.

The presence of up to 0.2 wt % fly ash in the atomized molten carbonate did not affect the removal of SO_2 from flue gas.

Fly ash settled from suspension in the unagitated molten carbonate.

Commercial nozzles with larger liquid passages will probably atomize ash-containing melts without plugging.

III. THEORETICAL ASPECTS OF SULFUR DIOXIDE ABSORPTION

In gas absorption by a spray of droplets there must be transfer of reactants from both phases to the reaction zone near the gas-liquid interface. The liquid-phase transfer of carbonate from the interior to the surface of a droplet is discussed in the following section. Next, the gas-phase mass transfer of sulfur dioxide to the surface of a falling droplet is developed and interpreted in light of spray absorption tests.

A. DIFFUSION WITHIN A SPRAY DROPLET

1. Introduction

The reduced effectiveness of SO_2 removal observed for the G/L ratio (SO_2 molar flow rate)/(Melt molar flow rate) less than unity suggests that diffusion within carbonate droplets may hinder absorption of SO_2.

Exact analysis of the absorption mechanism would be difficult since it is complicated by such phenomena as

a. Augmented mass transfer by droplet oscillation

b. Time-varying ambient SO_2 concentration at the droplet surface

c. Some dissolution, diffusion into, and reaction of SO_2 within the droplet interior.

However, a simple physical model can be formulated and analyzed to predict the approximate time required for $CO_3^=$ ion diffusion from a droplet interior to the surface.

2. The Physical Model

Assume that the reaction

$$M_2CO_3 + SO_2 \rightarrow M_2SO_3 + CO_2 \qquad (10)$$

occurs at the droplet surface so rapidly that the carbonate concentration $[CO_3^=]$ at the droplet surface r = R, is virtually zero or

$$[CO_3^=]_{r=R} = 0 \qquad (11)$$

Because one mole of $SO_3^=$ is formed for each mole of $CO_3^=$ reacted and the densities of M_2CO_3 and M_2SO_3 are about equal, there is no appreciable convective flux (net velocity of salt with respect to the droplet center) and the droplet radius R is constant.

Molecular diffusion at time t within a sphere is described by the equation

$$\frac{\partial c}{\partial t} = \mathbb{D}\left(\frac{1}{r^2}\ \frac{\partial}{\partial r}\ \left(r^2\ \frac{\partial c}{\partial r}\right)\right) \qquad 0 \leq r \leq R \qquad (12)$$

Where $\mathbb{D}$ and c are respectively the diffusivity and concentration of $CO_3^=$ ions in M_2CO_3. Boundary conditions pertinent to the model are

$$c = c_o, \quad t < 0, \; 0 \leq r \leq R$$

$$\frac{\partial c}{\partial r} = 0, \quad r = 0$$

$$c = 0, \quad r = R, \; t \geq 0 \qquad (13)$$

3. Analysis of the Model

The above boundary value problem is analogous to the heat combustion boundary value problem described by Carslaw and Jaeger[7]. The average concentration of $CO_3^=$ ions in the droplet is related to time and droplet radius by

$$\frac{c}{c_o} = \text{function of } (\mathbb{D}t/R^2) \qquad (14)$$

where c_o is the initial concentration of M_2CO_3. Using the value $\mathbb{D} = 1.06 \times 10^{-5}$ cm^2/ sec at 420°C (788°F)[8] the following percent reaction values shown in Table 11, are obtained for M_2CO_3 droplets of various sizes.

4. Discussion

It is evident that, for the physical model described, M_2CO_3 is well utilized in a fine spray (droplets smaller than 100 microns) and contact times greater than 0.3 seconds.

The maximum residence time of droplets in the 15-cm test column studies, with gas velocities of 104 ft/min was (3 ft column length)/(104 ft/min) = 0.029 minutes = 1.73 sec. Small droplets probably traveled only half of the column length since the nozzle was located in the middle of the column. Furthermore, droplets were readily collected on the column walls. An average droplet residence time of the order of 0.3 sec is therefore reasonable. Samples of exit melt analyzed for M_2CO_3 content averaged 0.43, 0.84, and 0.98 mole fraction M_2CO_3, depending on melt to SO_2 flow rate ratio. The concentration of M_2CO_3 observed in the exit melt, when compared with theoretical values in the foregoing table, suggest that droplet size in the spray was larger

Table 11. Extent of Carbonate Reaction in Droplets for Various Droplet Diameters and Times of Contact

Droplet diameter, 2R (microns)	Time of contact (seconds)	Dt/R^2	Percent of M_2CO_3 reacted, $100(1-C/C_o)$
10	0.1	4.24	100
32		0.424	99
100		0.042	56
320		0.004	20
10	0.3	12.72	100
32		1.27	100
100		0.13	83
320		0.01	30
10	1.0	42.4	100
32		4.24	100
100		0.42	99
320		0.04	53
10	3.0	127.2	100
32		12.7	100
100		1.27	100
320		0.13	83

than about 300 microns.

B. MASS TRANSFER TO A POLYDISPERSE AEROSOL OF LIQUID DROPLETS

Within a spray reactor droplets of liquid are atomized into a rising stream of gas. Very small droplets rise with the gas as would fog, while very large droplets fall rapidly, like raindrops. However, each droplet falls at its terminal velocity v_t relative to the gas. Mass transfer of SO_2 to a droplet is a function of Reynolds number and diffusivity of SO_2 in its carrier gas.

No two drops generated by a nozzle are the same size. One cannot specify a priori an "effective" or "equivalent" drop diameter. Each droplet must be considered individually or, more practically, droplets are assigned to size classes for calculation.

The number of droplets of diameter d_i per cubic centimeter of spray such that $(d_i - \frac{1}{2}\Delta d) < d_i < (d_i + \frac{1}{2}\Delta D)$ is expressed by

$$\#i = \frac{\left\{\frac{\text{gm liquid sprayed}}{\text{sec}}\right\}\left\{\begin{matrix}\text{fraction of liquid forming droplets}\\ \text{of diameter } d_i \pm \frac{1}{2}\Delta D\end{matrix}\right\}}{\left\{\begin{matrix}\text{volume of a droplet}\\ \text{of diameter } d_i\end{matrix}\right\}\left\{\frac{\text{gm liquid}}{\text{cm}^3}\right\}\left\{\begin{matrix}\text{velocity of droplets relative}\\ \text{to the absorber walls}\end{matrix}\right\}}$$

$$= \frac{6LM\, f(d)\Delta D}{\pi \rho\, d_1^3\, |v_1|} \qquad (15)$$

where

L = molal flux of liquid, moles liquid/cm^2/sec

M = molecular weight of liquid, gm liquid/mole

ρ = liquid density, gm/cm^3

and f(d) is the droplet size distribution function defined such that the fraction of liquid mass contained in droplets of diameter $d_i \pm \frac{1}{2}\Delta D$ equals $f(d_1)\Delta d$.

Consider a disc-shaped control volume bounded by the absorber walls and planes Δz cm apart, each perpendicular to the absorber axis. A mass

balance of SO_2 entering, leaving and reacting within the control volume gives

$$AG_{SO_2}\Big|_z - AG_{SO_2}\Big|_{z+\Delta z} - \Sigma r_i \, A\Delta z = 0 \tag{16}$$

Where A is the column's cross-section area, G_{SO_2} is the SO_2 flux (moles/cm^2/sec) and r is the per-unit-volume rate at which SO_2 is absorbed by droplets of the i^{th} size class. As $\Delta z \to 0$

$$\frac{d}{dz} G_{SO_2} + \Sigma r_i = 0 \tag{17}$$

$$U \frac{d}{dz} C_{SO_2} + \Sigma r_i = 0 \tag{18}$$

where U is the gas velocity and C_{SO_2} is the molar concentration of SO_2.

A single droplet of diameter d_i falling at velocity v_{ti} relative to the gas phase absorbs SO_2 according to the correlation[9]

$$\frac{k\, d_i}{C_f \mathcal{D}_f} = 2.0 + 0.60 \left[\frac{d_i \, v_{ti}}{\nu_f}\right]^{\frac{1}{2}} \left[\frac{\nu}{\mathcal{D}}\right]_f^{\frac{1}{3}} \tag{19}$$

The subscript denotes film properties. The Schmidt number $\frac{\nu}{\mathcal{D}}$ is essentially unity for $SO_2 - N_2$ and can be neglected. C is total gas concentration (gm gas/cc) and since the film is assumed isothermal and because the SO_2 concentration is low, $C_f = C$. The symbol k denotes the mass transfer coefficient. The rate of SO_2 absorption per droplet ($\dot{r}$) for these conditions is given by

$$\dot{r}_i = -k_i \pi d_i^2 \frac{0 - X_{SO_2}}{1 - X_{SO_2}} \text{ gm mole } SO_2/\text{ sec/droplet} \tag{10}$$

Since X_{SO_2} (X denotes mole fraction) is small ($X_{SO_2} \ll 1$) one may write

$$\dot{r}_i = k_i \pi d_i^2 X_{SO_2} \tag{20}$$

The per-unit-volume rate of SO_2 depletion is then (with $C_{SO_2} = C\ X_{SO_2}$)

$$r_i = \dot{r}_i f_i \tag{21}$$

Combining equations (19), (20), and (21) gives

$$\frac{d}{dz} C_{SO_2} + \left\{ \frac{\pi D}{U} \sum_i \frac{6LMf(d_i)\Delta D}{\pi \rho d_i^3 |v_i|} d_i \left[2.0 + 0.60 \frac{(d_i |v_{ti}|)}{\nu} \right]^{\frac{1}{2}} \right\} C_{SO_2} = 0 \tag{22}$$

From the equation form defining the HTU, $\frac{dc}{dz} + \frac{C}{HTU} = 0$, the HTU for the absorber is seen to be

$$HTU = \frac{\rho U / 6 D L M}{\sum_i \frac{2.0 + 0.6(d_i |v_{ti}|/\nu)^{\frac{1}{2}}}{d_i^2 |v_i|} f(d_i)\ \Delta D} \tag{23}$$

The preceding expression applies separately to the upper and lower sections of the absorber for which the droplet size distributions are different.

The data describing the droplet size distribution for the Spraying Systems Co. Whirljet nozzle fit well to a log-normal distribution for which the geometric standard deviation is $\sigma_g = 1.455$. The values of $f(d)\Delta D$ and HTU in Table 12 were calculated for distribution with mass median diameters of d = 300 and 500 microns.

Table 12 Evaluation of HTU for Polydisperse Spray Absorber

$$G_{25\ ft/sec} = \left(760 \frac{cm}{sec}\right)\left(4.90 \times 10^{-4} \frac{gm}{cm^3}\right)\left(3 \times 10^{-3} \frac{\text{mole } SO_2}{\text{mole gas}}\right)/29 \frac{\text{gm gas}}{\text{mole gas}}$$

$$= 3.84 \times 10^{-5} \frac{\text{mole } SO_2}{cm^2\ sec}$$

$$L/G = 4 \qquad L = 1.54 \times 10^{-4} \frac{\text{mole } M_2CO_3}{cm^2\text{-sec}}$$

Table 12, cont.

$$\frac{\rho_{liq}\, U_{gas}}{60 LM} = \frac{(1.97\ \frac{gm\ M_2CO_3}{cm^3})(760\ \frac{cm}{sec})}{6(0.69\ \frac{cm^2}{sec})(1.54 \times 10^{-4}\ \frac{mole\ M_2CO_3}{cm^2\ sec})(100\ \frac{g\ M_2CO_3}{mole\ M_2CO_3})} = 23,500\ \frac{sec}{cm^2}$$

$$HTU = \frac{\rho\, U}{60 LM} \Big/ \sum \frac{2.0 + 0.6 \left(\frac{d_i |v_{ti}|}{\nu}\right)^{\frac{1}{2}}}{d_i^2 |v_i|}\ f(d_i)\Delta d$$

CALCULATION OF SUMS

	d_i μ	$f(d_i)\Delta d$	v_{ti} cm/sec	$\frac{d_i v_{ti}}{\nu}$ Re_i	$2 + 0.6\ Re^{\frac{1}{2}}$	v_i cm/sec	$\frac{2 + 0.6\ Re^{\frac{1}{2}}}{d_i^2\ v_i}$
Mass Median Diameter = 300 μ	100	0.07	30	.45	2.40	730	32.9
	200	0.14	108	3.2	3.07	652	11.8
	250	0.19	140	5.3	3.38	620	8.2
	300	0.18	170	7.6	3.65	590	6.9
	350	0.15	200	10.5	3.94	560	5.7
	410	0.13	240	14.7	4.30	500	5.1
	500	0.09	310	23	4.88	450	4.3
	600	0.03	390	35	5.55	370	4.2
	700	0.01	450	47	6.11	310	4.0
	800	0.01	560	67	6.92	200	5.0
Mass Median Diameter = 500 μ	200	0.03	108	1.79	3.07	652	11.8
	285	0.09	160	2.59	3.35	600	7.3
	350	0.10	200	3.24	3.94	560	5.7
	400	0.11	236	3.75	4.23	524	5.1
	450	0.12	266	4.21	4.53	494	4.5
	500	0.10	310	4.80	4.88	450	4.3
	550	0.09	336	5.48	5.29	424	4.1
	610	0.12	384	5.91	5.55	376	3.96
	700	0.11	450	6.85	6.11	310	4.0
	800	0.06	560	8.2	6.92	200	5.0
	920	0.04	608	9.13	7.48	152	5.8
	1050	0.014	710	10.4	8.25	50	15.0

CALCULATION OF HTU

	Geometric Mean Droplet Diameter	
	300 microns	500 microns
$\sum_1 \frac{2.0 + 0.6\ N_{Re}^{\frac{1}{2}}}{d_i^2 \lvert v_i \rvert} f(d_i)\Delta d$	8.88	5.09
$HTU = \frac{\rho U / 6 \mathscr{D} LM}{\Sigma}$	24.5 meters	44.0 meters

The HTU's calculated in the foregoing illustration are exceedingly high and would allow only about 4% removal of SO_2 in a one-meter-long column. But SO_2 removals in the experimental spray column were greater than 90%. It follows then that some other mechanism, not accounted for in the foregoing example, must operate.

Inspection of Equation 23 shows that the contribution to the HTU for droplets of diameter d_i varies with $|v_i|/\sqrt{|v_i|}$ when $0.6\ (d_i|v_{ti}|/\nu)^{\frac{1}{2}} \gg 2$. The condition for which $|v_i|/\sqrt{|v_i|}$ is decreased appreciably, a necessary criterion to minimize the HTU, is that of droplets being injected into the oncoming stream of gas. In this situation v_i, the velocity of the droplet relative to the reactor walls quickly reaches zero as the oncoming gas stream blows it back. Similarly v_{ti}, the velocity of the droplet relative to the gas, is initially greater than the terminal velocity of the droplet by gravity only. But, such conditions, as in the experiments, occur only in the vicinity of the outlet of a nozzle sprayed into a gas stream.

It is expected that in the test column v_i, velocity of droplets relative to the reactor walls, assumed initial values of about -200 cm/sec initially, increased to through zero and finally reached about 760 cm/sec. Similarly v_{ti}, velocity of droplets relative to the gas, decreased from about -1000 cm/sec initially to the relaxed terminal velocity of the droplets, -170 and -310 cm/sec for 300 and 500 droplets. Other conditions favorable to mass transfer not accounted for in the physical model, probably occurred in the test column. These include turbulent fluctuation of the gas stream which increase v_t, and oscillation of liquid droplets.

The above discussion leads to the important conclusion that absorption of gas by a spray occurs near the spray generating nozzle where the initial droplet velocity is large. When droplets slow to their terminal velocity due to gravity, mass transfer is reduced almost to insignificance. Johnstone and Kleinschmidt (5) concluded from their inspection of spray data that..."the rate of absorption was high immediately in front of the nozzles but that it dropped off rapidly with distance. While the size distribution of the droplets from the nozzles was not known, it is obvious that the smallest particles, which were the source of the major portion of the absorption, lost their initial kinetic energy and could not penetrate more than a few feet into the gas."

The preceding discussion and conclusions suggest the form of an absorption reactor for the Small Pilot Plant. The reactor need be only several feet high, allowing about two feet between planes of nozzles, and space above for a demister. The recommended absorber design values are listed in Table 13.

Table 13. Recommended Design Values for the Pilot Plant Absorber

Power equivalent of system	0.66 MWe
Rate of SO_2 formation	0.6 lb mole/hr
Gas velocity in absorber	25 ft/sec (760 cm/sec)
SO_2 concentration in gas	0.3 mole percent
Temperature	450°C (850°F)
SO_2 flux (G)	3.86×10^{-5} $\frac{\text{g-mole } SO_2}{\text{cm}^2/\text{sec}}$
G/L, molar gas to liquid ratio	0.2 maximum (minimum L/G = 5)
Carbonate flux (L) molar	1.93×10^{-4} $\frac{\text{g-mole } M_2CO_3}{\text{cm}^2/\text{sec}}$ minimum
volumetric	0.145 gallon/ft^2/min minimum
	4.0 gallon/ft^2/min maximum

Table 13, cont.

Gas pressure drop through column, 20 in. diameter, 20 ft length, at minimum liquid spray flux (calculations in Notebook 4458-1)

Pipe friction	37 dyne/cm^2
Suspension of spray	0.2 dyne/cm^2
Reversal of droplet motion	15.5 dyne/cm^2
Total (demister not included)	52.7 dyne/cm^2 = 0.021 in. H_2O

Nozzle arrangement: Three planes of seven nozzles each, planes spaced 2 ft apart, uppermost plane set in a honeycomb of seven cells for spray collection and wetted-wall effect

Nozzle type:

Lower plane	Pneumatic
Middle plane	Whirljet
Top plane	Whirljet

Demister: Placed 24 in. above the top plane of nozzles. Type unspecified. Removable. Contributions of demister to pressure drop and absorber efficiency to be determined experimentally.

APPENDIX A. The Relation Between Transfer Unit Height (HTU) and Reynolds Number in a Wetted-Wall Column.

A mass balance about a disc-shaped control volume of thickness Δz and face area equal to that of the wetted-wall column,

$$\frac{\pi D^2}{4} VCX\Big|_z - \frac{\pi D^2}{4} VCX\Big|_{z+\Delta z} - \pi D \Delta z k_g X = 0 \qquad (A1)$$

gives in the limit as $\Delta z \to 0$

$$\frac{dX}{dz} + \frac{X}{CDV/4k_g} = 0 \qquad (A2)$$

The quantity $CDV/4k_g$ is, by definition, the gas phase HTU

In the above expressions:

d = column diameter cm

V = mean gas velocity cm/sec

z = distance variable along column axis cm

X = mole fraction of SO_2 in gas

C = molar density of gas moles/cm^3

k_g = mass transfer coefficient moles/cm^3/sec

The Sherwood Number is defined as

$$N_{SH} = \frac{k_g D}{C \mathcal{D}} \qquad (A3)$$

By substitution in Equation A2

$$HTU = \frac{D^2}{4 \mathcal{D} N_{SH}} \qquad (A4)$$

For the systems SO_2-CO_2 and CO_2-N_2, the Schmidt number, N_{sc}, is within 5% of unity whereby one may write the Gilliland-Sherwood Equation

$$N_{SH} = 0.023 \left(\frac{Dv}{\nu}\right)^{0.83} \left(\frac{\nu}{\mathcal{D}}\right)^{0.44} \tag{A5}$$

as

$$N_{SH} = 0.023 \left(\frac{Dv}{\nu}\right)^{0.83} \left(\frac{\nu}{\mathcal{D}}\right)^{1.0} \tag{A6}$$

Combining Equations A4 and A6 gives

$$HTU = \frac{D^2 v}{4\,\mathcal{D}\,0.023} \left(\frac{Dv}{\nu}\right)^{0.83} \left(\frac{\nu}{\mathcal{D}}\right) \tag{A7}$$

which simplifies to

$$HTU = \frac{D \left(\frac{Dv}{\nu}\right)^{0.17}}{4\,(0.023)} \tag{A8}$$

or

$$HTU = 11\ D\ N_{Re}^{0.17} \tag{A9}$$

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